Divergence of the Heavy Quasiparticle Mass at the Antiferromagnetic Quantum Critical Point in YbRh₂Si₂*

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We report low temperature specific heat, C, magnetization, M, susceptibility, χ , and electrical resistivity, ρ , measurements on high-quality single crystals of the heavy-fermion system YbRh₂(Si_{1-x}Ge_x)₂ (x=0 and 0.05). The undoped compound shows weak antiferromagnetic (AF) order at $T_N=70$ mK which is suppressed to below 10 mK by a tiny volume expansion in the x=0.05 system. In the latter pronounced deviations from Landau Fermi liquid (LFL) behavior occur, e.g. $\Delta \rho \sim T$ over three decades in T. Both thermodynamic and magnetic properties show a crossover at about 0.3 K: At 0.3 K $\leq T \leq 10$ K we observe $C/T \sim \log(T_0/T)$ and a "non-Curie" behavior $\chi^{-1} \sim T^{\alpha}$ with $\alpha < 1$ similar to what was found for the prototypical system CeCu_{5.9}Au_{0.1}. Below 0.3 K, χ turns into a Curie-Weiss dependence $\chi^{-1} \sim (T-\Theta)$ indicating large unscreened Yb³⁺ moments whereas in C(T)/T a pronounced upturn occurs.

In the undoped compound the AF order is suppressed continuously by critical fields $B_{c0} \simeq 0.06$ T and 0.7 T applied perpendicular and parallel to the c-axis, respectively. For $B > B_{c0}$ a LFL state with $\Delta \rho = A(B)T^2$ and $C(T)/T = \gamma_0(B)$ is induced, that fulfills the Kadowaki-Woods scaling $A \sim \gamma_0^2$. Upon reducing the magnetic field to B_{c0} a $1/(B - B_{c0})$ dependence of A(B) and $\gamma_0^2(B)$ indicates singular scattering at the whole Fermi surface and a divergence of the heavy quasiparticle mass.

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1. Introduction

The origin of non-Fermi liquid (NFL) behavior in heavy-fermion (HF) systems has been studied intensively in the past decade but is still unclear up to now [1]. In particular two different scenarios are discussed for the quantum-critical point (QCP), where long-range antiferromagnetic (AF) order emerges from the HF state; one in which NFL behavior arises from Bragg diffraction of the electrons off a critical spin-density wave (SDW) [3, 4, 5], the other in which the bound-state structure of the composite heavy fermions breaks up at the QCP resulting in a collapse of the effective Fermi temperature [2, 6]. In the SDW scenario, assuming threedimensional (3D) spinfluctuations, singular scattering occurs only along certain "hot lines" connected by the vector **q** of the near AF order while the remaining Fermi surface still behaves as a Fermi liquid. Therefore, the lowtemperature (T) specific heat coefficient C(T)/T, that measures the heavy quasiparticle (QP) mass, is expected to show an anomalous temperature dependence $C(T)/T = \gamma_0 - \alpha \sqrt{T}$, but remains finite at the QCP [5]. A diverging QP mass, as evident from the $C(T)/T \sim \log(T_0/T)$ behavior found e.g. in the prototypical system $CeCu_{6-x}Au_x$ for $x_c = 0.1$ [7], would arise only if truly 2D critical spinfluctuations render the entire Fermi surface "hot" [8]. On the other hand, measurements of the inelastic neutron scattering on CeCu_{5.9}Au_{0.1} [9] showed that the critical component of the spin fluctuations is almost momentum independent leading to the proposal of the locally critical scenario [6, 9]. Since T-dependent measurements at the QCP alone provide no information on how the heavy quasiparticles decay into the quantum critical state it is necessary to tune the system away from the magnetic instability in the Landau Fermi liquid (LFL) state and to follow the QP properties upon approaching the QCP. In this paper we demonstrate that magnetic fields can be used for this purpose.

In the following, we consider the HF metal YbRh₂Si₂ for which pronounced NFL phenomena, i.e., a logarithmic divergence of C(T)/T and a quasi-linear T-dependence of the electrical resistivity below 10 K, have been observed above a low-lying AF phase transition [10]. This system is very suitable for such an investigation, because the effect of disorder is negligible in crystals with a residual resistivity of less than 1 $\mu\Omega$ cm. The application of pressure to YbRh₂Si₂ increases T_N [10] as expected, because the ionic volume of the magnetic $4f^{13}$ Yb³⁺-configuration is smaller than that of the nonmagnetic $4f^{14}$ Yb²⁺ one. Expanding the crystal lattice by randomly substituting Ge for the smaller isoelectric Si atoms allows one to tune YbRh₂(Si_{1-x}Ge_x)₂ through the QCP without affecting its electronic properties and without introducing significant disorder to the lattice [11]. In YbRh₂(Si_{1-x}Ge_x)₂ with the nominal Ge concentration x = 0.05 (see below) the NFL behavior

extends to the lowest accessible temperatures, in particular $\Delta \rho(T) \sim T$ is observed from above 10 K down to 10 mK [11].

This article is organized as follows: After giving details concerning experimental techniques in the next section, we address in section 3 the pronounced NFL behavior at zero magnetic field in thermodynamic, magnetic and transport properties at the QCP in YbRh₂(Si_{0.95}Ge_{0.05})₂. In section 4 we concentrate on the undoped system and prove that magnetic fields can be used to tune the system continuously from the AF ordered state through the QCP into a field-induced LFL state. We present evidence for the divergence of the heavy quasiparticles at the QCP and end with the conclusions in section 5.

2. Experimental details

Single crystalline platelets of YbRh₂(Si_{1-x}Ge_x)₂ prepared with the nominal Ge compositions x = 0 and 0.05 were grown from In flux as described earlier [10, 11]. A microprobe analysis (MA) of the x = 0.05 sample revealed a Ge concentration $x_{MA} \leq 0.02$, only. The solubility of Ge atoms in In flux is much higher compared to that of Si atoms. This causes the actual Ge concentration being smaller than 0.05 in these single crystals. To be consistent with previous publications [11, 12], the Ge-doped single crystals studied in this paper are labeled by their nominal Ge composition x = 0.05. To demonstrate that the main effect of Ge doping is primarily the expansion of the volume, measurements of the electrical resistivity under hydrostatic pressure have been performed on a x = 0.05 single crystal [11, 13]. At a pressure of 0.63 GPa the onset of AF order has been observed at $T_N = 0.185$ K. Furthermore, the T_N vs p phase diagram of the Ge-doped crystal matches perfectly with that found for pure YbRh₂Si₂ [10] if the pressure axis is shifted by -0.2 GPa [13]. Taking the value $B = (198 \pm 15)$ GPa for the bulk modulus as determined from recent Moessbauer measurements under hydrostatic pressure [14], this pressure shift corresponds to a volume expansion of 0.1% for the Ge-doped sample consistent with a Ge content of 0.02 ± 0.004 .

The new generation of x=0 crystals show a residual resistivity $\rho_0 \simeq 1\mu\Omega$ cm, i.e., twice as low as ρ_0 of the previous ones. Whereas for the latter the phase transition at T_N discovered by AC-susceptibility measurements could not be resolved in the resistivity [10], the new crystals show a clear kink of $\rho(T)$ at T_N , see below.

For all low-temperature measurements, ${}^{3}\text{He}/{}^{4}\text{He}$ dilution refrigerators were used. The specific heat was determined with the aid of a quasi-adiabatic heat pulse technique. The electrical resistivity, ρ , and the magnetic AC-susceptibility, χ , were measured utilizing a Linear Research Co. (LR700)

bridge at 16.67 Hz. Amplitudes of 0.1 mA and 1 Oe for the current and magnetic field, respectively, were chosen to determine ρ and χ . Absolute values of χ have been determined from a comparison in the temperature range 2 K $\leq T \leq$ 6 K with the results of the dc-susceptibility in 50 mT using a Quantum Design SQUID magnetometer. Low temperature magnetization measurements were performed utilizing a high-resolution capacitive Faraday magnetometer as described in [15].

3. Zero field NFL behavior in YbRh₂(Si_{0.95}Ge_{0.05})₂

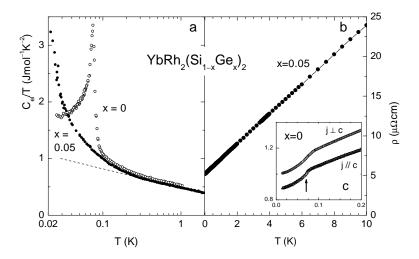


Fig. 1. Comparison of the low-temperature electronic specific heat $C_{el} = C - C_Q$ as C_{el}/T vs $\log(T)$ (a) and the electrical resistivity ρ (b and c) of high-quality YbRh₂(Si_{1-x}Ge_x)₂ single crystals with Ge-contents x=0 and x=0.05. $C_Q \sim T^{-2}$ is the nuclear quadrupole contribution calculated from recent Mössbauer results [14]. Dotted line in (a) marks $\log(T_0/T)$ dependence with $T_0 \simeq 24$ K. Resistivity data shown in (c) were obtained for the current **j** flowing along (Δ) and perpendicular (o) to the c-axis. The arrow indicates T_N as obtained from the maximum in $d\rho/dT$.

In undoped YbRh₂Si₂ we observe a second order phase transition in specific heat (Fig. 1a) which, according to AC-susceptibility measurements (inset of Fig. 2), marks the onset of AF order [10]. The resistivity follows a quasi-linear T-dependence down to about 80 mK, below which a sharp decrease, independent of the current direction, is observed (inset of Fig. 1b). Thus the resistivity does not show any signatures of the formation of a SDW for which an increase of $\rho(T)$ along the direction of the SDW modulation,

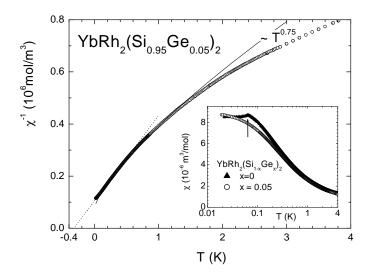


Fig. 2. Inverse of the AC-susceptibility in the easy-magnetic plane perpendicular to the crystallographic c-direction, as χ^{-1} vs T for YbRh₂(Si_{0.95}Ge_{0.05})₂. The dotted line indicates Curie-Weiss type dependence $\chi(T) = C/(T-\Theta)$ implying large fluctuating moments of 1.4 μ_B/Yb^{3+} -ion and a Weiss temperature of $\Theta \simeq -0.3$ K. The solid line represents $\chi^{-1} \sim T^{\alpha}$ with $\alpha \approx 0.75$ (see text). The inset shows χ vs T on a logarithmic scale for YbRh₂(Si_{1-x}Ge_x)₂ for x = 0 and 0.05. The arrow indicates T_N for the undoped system.

indicating the partial gapping of the Fermi surface, should be expected. The absence of this behavior favors the interpretation of local-moment type of magnetic order in YbRh₂Si₂. The resistivity in the AF ordered state (at B=0) is best described by $\Delta \rho = AT^2$ with a huge coefficient, $A=22~\mu\Omega$ cm/K², for 20 mK $\leq T \leq 60$ mK. Extrapolating $C_{el}(T)/T$ as $T \to 0$ to $\gamma_0 = (1.7 \pm 0.2) \text{ J/K}^2$ mol reveals an entropy gain at the AF phase transition of only about $0.03R \cdot \ln 2$. This provides evidence for the weakness of the AF order in YbRh₂Si₂. The ratio of A/γ_0^2 in the ordered state is close to that expected for a LFL [16], i.e., one with very heavy quasiparticle masses. The AF ordering is suppressed to below 10 mK by doping with a small amount of Ge in $YbRh_2(Si_{0.95}Ge_{0.05})_2$. This system must be located very close to the QCP, and pronounced NFL behavior is observed down to lowest T: The electronic specific heat C_{el}/T follows a $\log(T_0/T)$ $(T_0 \simeq 24 \text{ K} [10])$ behavior between 0.3 and 10 K. In the same T-range a linear T-dependence of the electrical resistivity is observed. Thus above 0.3 K (e.g. $0.0125 T_0$) resembling T-dependences as reported for CeCu_{5.9}Au_{0.1} are observed that could only be explained within the SDW scenario assuming truly 2D critical

spinfluctuations [8]. Below that temperature, which corresponds to 75 mK in CeCu_{5.9}Au_{0.1} ($T_0 = 6$ K), a cross-over into a different regime takes place. Whereas the resistivity continuous to follow a linear T-dependence down to below 15 mK, a pronounced upturn is observed in C_{el}/T whose origin will be discussed in the conclusion. A cross-over around 0.3 K is observed in the susceptibility, too: Below 0.3 K, χ follows a Curie-Weiss type behavior (Fig. 2) implying fluctuating moments of the order of 1.4 $\mu_B/$ Yb³⁺-ion. The Weiss-temperature of $\Theta \simeq -0.32$ K suggests strong AF correlations in this regime. Above 0.3 K, the susceptibility can be described by $\chi^{-1} \sim (T^{\alpha} - \Theta)$ with the exponent α decreasing continuously from 1 (below 0.3 K) to 0.5 at 2 K. As displayed in Fig. 2, in the T interval 0.2 K $\leq T \leq$ 1.4 K the susceptibility roughly follows $\chi^{-1} \sim (T^{\alpha} - \Theta)$ with $\alpha \approx 0.75$ and $\Theta \approx -0.05$ K, i.e. a "non Curie-Weiss" behavior reminiscent of CeCu_{5.9}Au_{0.1} [9].

4. Field tuning YbRh₂Si₂ through the QCP

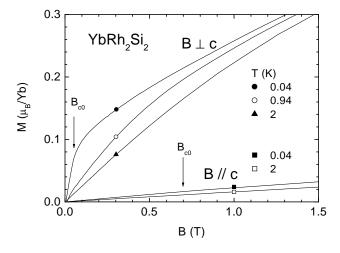


Fig. 3. Isothermal DC magnetization M at varying temperatures in magnetic fields applied along and perpendicular to the c-axis. Arrows indicate critical fields B_{c0} .

As discussed in the previous section, the weak AF order in YbRh₂Si₂ can be suppressed by a tiny volume expansion upon replacing Si by larger Ge-atoms, and pronounced deviations from LFL behavior occur at the QCP in YbRh₂(Si_{0.95}Ge_{0.05})₂. To study how the heavy quasiparticles decay into the quantum critical state it is required to investigate their properties upon crossing the QCP at zero temperature as a function of a control parameter that can be varied continuously. In the following we will show that the application of magnetic fields to YbRh₂Si₂ can be used for this purpose.

This study was motivated by the observation of field-induced NFL behavior in the doped AF systems $\text{CeCu}_{6-x}\text{Ag}_x$ [17, 18] and $\text{YbCu}_{5-x}\text{Al}_x$ [19]. YbRh_2Si_2 is highly suited to study the properties of a B-induced QCP, because the influence of disorder in this clean stoichiometric system should be negligible. Furthermore, the ordering temperature $T_N = 70$ mK is the lowest among all undoped HF systems at ambient pressure and already a very small critical magnetic field $B_c(0) = B_{c0}$ is sufficient to push T_N towards zero temperature.

We first discuss the low-temperature magnetization which proves that the AF phase transition as a function of field is a continuous one. YbRh₂Si₂ exhibits a highly anisotropic magnetic response, indicating that Yb³⁺ moments are forming an "easy-plane" square lattice perpendicular to the crystallographic c-direction [10]. The isothermal magnetization (Fig. 3) shows a strongly nonlinear response for fields $B \perp c$. For $T < T_N$ a clear reduction in slope is observed above 0.06 T which indicates the suppression of AF order resulting in a weakly polarized state. A smooth extrapolation of M(B)for B > 0.06 T towards zero field reveals a value of $\mu_s < 0.1 \mu_B$ for the staggered magnetization in the AF state, indicating that the size of the ordered moments is much smaller than that of the effective moments observed in the paramagnetic state above T_N . Thus a large fraction of the local moments appears to remain (quantum) fluctuating within the easy plane in the AF ordered state. Their continuous polarization for fields exceeding B_{c0} gives rise to a strong curvature in M(B) for $B \perp c$. For fields applied along the magnetic hard direction, $B \parallel c$, the magnetization shows an almost linear behavior (Fig. 1b) which was found to extend at least up to 58 T [12]. At $T < T_N$ a very tiny increase in the M(B) slope is observed below about 0.7 T which, according to the resistivity measurements discussed below, represents the critical field B_{c0} for $B \parallel c$.

In Fig. 4 we show the evolution of the low-temperature resistivity upon applying magnetic fields along and perpendicular to the easy magnetic plane. At small magnetic fields the Néel temperature, determined from the maximum value of $d\rho/dT$, shifts to lower temperatures and vanishes at a critical magnetic field B_{c0} of 0.06 T in the easy magnetic plane and of 0.66 T along the magnetic hard direction, i.e. the crystallographic caxis. At $B = B_{c0}$, the resistivity follows a linear T-dependence down to the lowest accessible temperature of about 20 mK. This observation provides striking evidence for field-induced NFL behavior at the critical magnetic fields applied along both crystallographic directions [20]. At $B > B_{c0}$, we find $\Delta \rho = A(B) \cdot T^2$ for $T \leq T^*(B)$, with $T^*(B)$ increasing and A(B) decreasing upon raising the applied magnetic field. The evolution of T_N and T^* as a function of B is shown in Fig. 5. The extremely low value of the critical field applied along the easy plane highlights the near degeneracy of

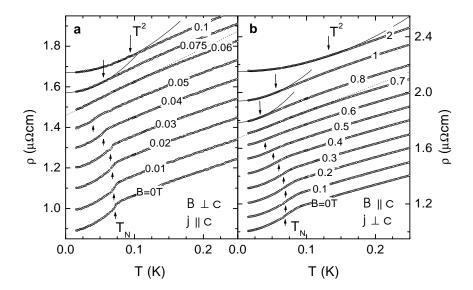


Fig. 4. Low-temperature electrical resistivity of YbRh₂Si₂ at varying magnetic fields applied perpendicular (a) and along the c-axis (b). For clarity the different curves in B>0 were shifted subsequently by 0.1 $\mu\Omega$ cm. Up- and downraising arrows indicate T_N - and upper limit of T^2 behavior, respectively. Dotted and solid lines represent $\Delta\rho \sim T^\epsilon$ with $\epsilon=1$ and $\epsilon=2$, respectively.

two different heavy LFL states, one being weakly AF ordered $(B < B_{c0})$ and the other one being weakly polarized $(B > B_{c0})$.

Next we address specific-heat measurements (Fig. 6) which have been performed at magnetic fields along the c-axis. Due to the strong magnetic anisotropy, the sample plate used for the specific-heat measurement could not be aligned perfectly along the hard magnetic direction. Therefore, a critical field B_{c0} of only about 0.3 T was sufficient to suppress AF order completely in these experiments. As shown in Fig. 6, at $B=B_{c0}$ the specific-heat coefficient $\Delta C(T)/T$ increases down to the lowest T, indicative of a field-induced NFL ground state. Within 40 mK $\leq T \leq$ 120 mK it follows a steep increase similar to the upturn observed for the x=0.05 single crystal below 0.3 K (c.f. Fig. 1a). While this anomalous contribution is strongly reduced upon increasing B, at magnetic fields $B \geq 1$ T, the nuclear contribution becomes visible at the lowest temperatures, above which a constant $\gamma_0(B)$ value is observed within a limited temperature window (Fig. 6). $\gamma_0(B)$ decreases in magnitude upon increasing the field.

A very similar behavior has been found in the field dependence of the low temperature AC susceptibility, too [10]. For magnetic fields exceeding B_{c0}

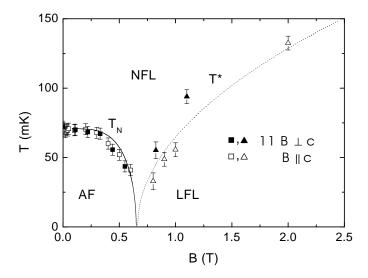


Fig. 5. T-B phase diagram for YbRh₂Si₂ with T_N as derived from $d\rho/dT$ vs T and T^* , the upper limit of the $\Delta \rho = AT^2$ behavior, as a function of magnetic field, applied both along and perpendicular to the c-axis. For the latter ones the B-values have been multiplied by a factor 11. Lines separating the antiferromagnetic (AF), non-Fermi liquid (NFL) and Landau Fermi liquid (LFL) phase are guides to the eye.

constant values $\chi_0(B)$ have been observed which decrease in size upon increasing B.

In Fig. 7 we present our analysis of the magnetic-field dependence of the coefficients A, γ_0 and χ_0 observed for $T \to 0$ in the resistivity, $\Delta \rho = A(B)T^2$, specific heat, $\Delta C/T = \gamma_0(B)$ [10], and magnetic AC-susceptibility, $\chi = \chi_0(B)$ [10] when approaching the QCP upon reducing B towards B_{c0} . As shown in the inset of Fig. 7, we observe $A \sim \gamma_0^2$, $A \sim \chi_0^2$ and thus also $\gamma_0 \sim \chi_0$, independent of the field orientation and for all B values exceeding B_{c0} . Like in the AF ordered state (at B=0) we find that the A/γ_0^2 ratio roughly equals that observed for many HF systems [16]. A very large Sommerfeld-Wilson ratio $R = (\chi_0/\gamma_0)(\pi^2 k_B^2/\mu_0 \mu_{eff}^2)$ of about 14 ($\mu_{eff} = 1.4\mu_B$) indicates a strongly enhanced susceptibility in the field-aligned state of YbRh₂Si₂ pointing to the importance of low-lying ferromagnetic ($\mathbf{q} = \mathbf{0}$) fluctuations, as also inferred from recent ²⁹Si-NMR measurements [21].

Since YbRh₂Si₂ behaves as a true LFL for $B > B_{c0}$ and $T < T^*(B)$, the observed temperature dependences should hold down to T = 0. The field dependence A(B) shown in Fig. 7 measures the QP-QP scattering cross section when, by field tuning, crossing the QCP at zero temperature. A

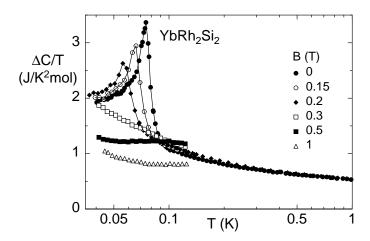


Fig. 6. Specific heat as $\Delta C/T = (C - C_Q)/T$ vs T (on a logarithmic scale) for YbRh₂Si₂ at varying fields applied parallel to the c-axis. $C_Q \sim T^{-2}$ is the nuclear quadrupole contribution calculated from recent Moessbauer results[14].

 $1/(B - B_{c0})$ divergence is observed indicating that the whole Fermi surface undergoes singular scattering at the *B*-tuned QCP. Most importantly, the relation $A \sim \gamma_0^2$ observed at elevated fields, suggests that also the QP mass diverges, i.e., as $1/(B - B_{c0})^{1/2}$, upon approaching B_{c0} .

5. Conclusion

The HF metal YbRh₂Si₂ shows a weakly antiferromagnetically polarized ground state which is suppressed to $T_N \to 0$ either by a small volume expansion in YbRh₂(Si_{0.95}Ge_{0.05})₂ or by the application of a critical magnetic field B_{c0} . At the QCP, pronounced deviations from LFL behavior are observed below 10 K in the specific heat and the electrical resistivity, in particular $C/T \sim \log(T_0/T)$ (above 0.3 K) and $\Delta \rho \sim T$ (down to the lowest T). According to the SDW scenario, such T-dependences would only arise assuming the scattering of truly 2D critical spinfluctuations.

In order to study the decay of the heavy quasiparticles in the quantumcritical state, we have used magnetic fields to suppress the AF order in YbRh₂Si₂. By field tuning the system through the QCP at a critical field B_{c0} one reaches a field-aligned state which at low temperatures can be described by the LFL model. The $1/(B - B_{c0})$ divergence of A(B) and $\gamma_0^2(B)$ indicates a divergence of the QP mass and QP-QP scattering cross section upon approaching the QCP. In the SDW model the parameter δ , given by the square of the inverse magnetic correlation length in the 2D spin fluid,

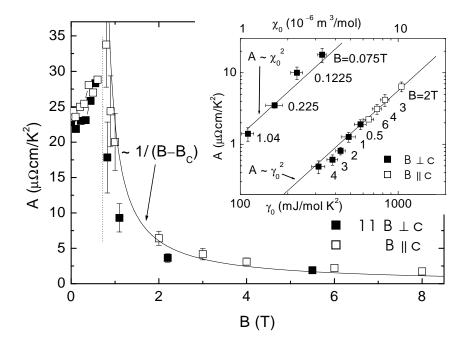


Fig. 7. Coefficient $A = \Delta \rho/T^2$ vs field B. Data for B perpendicular to the c-direction have been multiplied by 11. Dashed line marks B_{c0} , solid line represents $(B - B_{c0})^{-1}$. Inset shows double-log plot of A vs γ_0 and A vs χ_0 for different magnetic fields. Solid lines represent $A/\gamma_0^2 = 5.8 \cdot 10^{-6} \mu \Omega \text{cm} (\text{Kmol/mJ})^2$ and $A/\chi_0^2 = 1.25 \cdot 10^{12} \mu \Omega \text{cm} \text{K}^{-2}/(\text{m}^3/\text{mol})^2$.

measures the distance from the QCP, i.e., $\delta \sim (B - B_{c0})$. Assuming that the spin fluid renders the entire Fermi surface "hot", the coefficient A diverges as $A \sim 1/\delta$, whereas for the specific heat coefficient γ_0 a much weaker divergence $\gamma_0 \sim log(1/\delta)$ is expected [23]. Thus, this model would predict the ratio A/γ_0^2 to diverge upon decreasing B instead of being constant. The constancy of the Kadowaki-Woods ratio when approaching the QCP rather favors the locally critical scenario [24].

A stronger than logarithmic divergence of the QP mass is also evident from our B=0 measurements of $C_{el}(T)/T$ for YbRh₂(Si_{0.95}Ge_{0.05})₂ below 0.3 K(Fig. 1a). The upturn cannot be explained assuming a nuclear contribution to the low-T specific heat. Furthermore a very similar behavior is observed for the volume thermal expansion coefficient β too: As found for C_{el}/T , above 0.3 K β/T follows a logarithmic T dependence [22]. Below that temperature an even stronger divergence has been observed, providing additional evidence for the electronic origin of the upturn in the specific-

heat coefficient. The Curie-Weiss behavior, observed in $\chi(T)$ in the same T-range, where the upturn in $C_{el}(T)/T$ occurs, hints to large unscreened fluctuating Yb³⁺ moments persisting down to the lowest T at the QCP. This strongly suggests a local nature of the critical fluctuations.

To conclude, the QP mass diverges faster than logarithmic upon approaching the QCP in YbRh₂Si₂ in contradiction to the SDW picture and suggests a locally critical scenario for this system.

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